Pumping vortex into a Bose-Einstein condensate of heteronuclear molecules

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Abstract. Heteronuclear molecules attract wide attention due to their permanent electric dipole moments. Analogous to atoms with magnetic dipoles, the existence of nonzero electric dipoles significantly enhance possibilities and mechanisms for control and design of quantum degenerate molecule systems with electric (E-) fields. This work proposes a vortex creation mechanism inside a condensate of heteronuclear molecules through the adiabatic flipping of the axial bias of an analogous E-field Ioffe-Pritchard trap (IPT), extending the original protocol due to Ishoshima, *et al.*, [Phys. Rev. A **61**, 063610 (2000)] for an atomic spinor condensate inside a magnetic (B-) field IPT. We provide both analytic proof and numerical simulations to illustrate the high fidelity operation of this vortex pump protocol. We hope our work provides stimulating experimental possibilities for the active investigations in quantum degenerate molecule systems.

1. Introduction

The impressive progress in atomic quantum gases can be largely attributed to technical advances in the control of atomic quantum states utilizing electromagnetic interactions. While laser cooling with near resonant light narrows down atomic momentum distribution, forced evaporation on trapped atoms favors spatial confinement into the quantum degenerate regime. Since the first successful experiment on atomic Bose-Einstein condensation, or the controlled production of many bosonic atoms into the same quantum ground state of a trapping potential, the field of atomic quantum gases has blossomed into one of the leading frontiers in physics, attracting ever increasing interest. The ultimate goal along this line of thought is to control the state of an arbitrary assembly of an interacting many body system, and either use it to simulate other physical systems or to perform quantum computations.

Significant efforts have been directed toward the above ambitious goal, with the immediate pursuit from more recent endeavors targeted at the generation, control, and study of cold molecules. These endeavors include important successes such as the BCS to BEC cross-over due to tunable interactions induced through B-field magnetic Feshbach resonances and the production of quantum degenerate heteronuclear molecules in specific low lying ro-vibrational states of their electronic ground states [1]. The latter advance is especially illuminating, highlighting the prospects for observing and utilizing anisotropic and long range interactions from the permanent electric dipoles.

Molecule quantum fluids bring in new possibilities due to their richer internal degrees of freedom, such as alignment and rotation [2], even after their hyperfine spins like those of atomic spinor quantum gases are neglected. In this short note, we propose a simple mechanism to generate a vortex state, relying on the internal degrees of freedom of a heternuclear molecule, through adiabatically flipping the axial bias of an external E-field Ioffe-Pritchard trap (IPT) [3]. The physics of our proposal can be analogously mapped onto the successful protocol first proposed [4] and demonstrated for magnetically trapped atomic condensates [5, 6, 7]. Our theory hopefully will lead to parallel experiments that begin to address new opportunities afforded by the molecular internal degrees of freedom.

This paper is organized as follows. First, we describe our model for dc E-field trapping of cold heteronuclear molecules based on the interaction between their permanent electric dipoles with a spatially varying static E-field IPT. This is analogous to the operation of the famous IPT for magnetic dipoles with an inhomogeneous B-field [3]. The heteronuclear diatomic molecule is modeled as a 3D rigid rotor [2]. We then discuss the eigen structure of a heteronuclear molecule inside a dc E-field and identify states capable of trapped by a local E-field minimum and their associated quantum numbers. Next, the rotational properties of such eigen states are studied, which helps to illustrate the proposed vortex pump mechanism based on the flipping of the axial bias E-field [4, 5, 6, 7, 8, 9]. Finally we conclude.

It is imperative to present this study because this analogy between E-field and

B-field and between molecular E-dipole and atomic B-dipole is not a simple one to one map. The magnetic dipole of an atom contains both an electronic and a nuclear component, respectively, proportional to the electronic spin ($\vec{S}$) and the nuclear spin ($\vec{I}$). Its projection onto the local B-field is approximately proportional to the projection of its hyperfine spin $\vec{F} = \vec{S} + \vec{I}$ when the B-field is weak. In contrast, the molecular dipole is a fixed constant and pointed along the molecule axis connecting the two nuclei. The internal rotational angular momentum of the molecule stays in the perpendicular plane of the molecule axis in the absence of an E-field. Despite these differences, our study finds that the Isoshima, *et al.*, protocol [4, 5, 6], originally developed for B-field trapped atoms or molecules, remains effective for heteronuclear molecules in a E-field IPT. Much of these comparative studies and analogies will become clear from the materials presented in the third and fourth sections.

2. Trapping a heteronuclear molecule with static electric field

We consider a diatomic molecule composed of two different atomic nuclei and described in terms of their center of mass $\vec{R}$ and relative coordinate $\vec{r}$. Neglecting the nuclear and electronic spins and their associated interactions, or assume we consider an eigen state of the above internal degrees of freedom, the remaining Hamiltonian reads

$$\begin{aligned}\mathcal{H} &= \frac{\mathbf{P}_R^2}{2M} + \frac{\mathbf{P}_r^2}{2\mu} - \vec{D}(\vec{r})\cdot\vec{E}(\vec{R}) \\ &= \frac{\mathbf{P}_R^2}{2M} - \frac{\hbar^2}{2\mu r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial}{\partial r}\right) + \frac{\mathbf{J}^2(\theta_r,\phi_r)}{2\mu r^2} - \vec{D}(\vec{r})\cdot\vec{E}(\vec{R}),\end{aligned} \tag{1}$$

where $\vec{E}(\vec{R})$ denotes the inhomogeneous static E-field, which is slowly varying over the molecule size of r, and $\vec{D}(\vec{r})$ is the permanent electric dipole moment operator for this specific electronic and internal spin eigen-state [2, 10, 11]. $M = M_1 + M_2$ and $\mu = M_1M_2/(M_1 + M_2)$ are, respectively, the total and the reduced mass of the two atoms making up the molecule.

We adopt the Bohn-Oppenheimer approximation to study the effective trapping of the molecule center of mass motion. Within this approximation, the molecular wave function for $\vec{R}$ and $\vec{r}$ is decomposed into the form $\Psi(\vec{r},\vec{R}) = \Phi(\vec{r})\psi(\vec{R})$, and we find

$$\left[-\frac{\hbar^2}{2\mu r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial}{\partial r}\right) + \frac{\mathbf{J}^2(\theta_r,\phi_r)}{2\mu r^2} - \vec{D}(\vec{r})\cdot\vec{E}(\vec{R})\right]\psi_{M_J}(\vec{r}) = \mathcal{V}_{M_J}(\vec{R})\psi_{M_J}(\vec{r}), \tag{2}$$

$$\left(\frac{\mathbf{P}_R^2}{2M} + \mathcal{V}_{M_J}(\vec{R})\right)\Phi_n(\vec{R}) = E^{(n)}_{M_J}\Phi_n(\vec{R}). \tag{3}$$

Now consider for a specific vibrational state, the Eq. (2) above then reduces to the simple form of a 3D rotator in a dc E-field with a rotational constant $B = \langle\hbar^2/2\mu r^2\rangle$. If we further assume the local E-field direction to be the quantization z-axis, then the Schrödinger equation for the internal part of the diatomic molecule becomes

$$\left[BJ^2 - DE(\vec{R})\cos\theta_r\right]\psi_{M_J}(\theta_r,\phi_r) = \mathcal{V}_{M_J}(\vec{R})\psi_{M_J}(\theta_r,\phi_r), \tag{4}$$

where we have neglected the dependence of the permanent dipole moment on the internuclear distance $\vec{r}$.

Some intuition can be gained for weak E-fields if a simple perturbation theory is adopted as is done in Ref. [11]. We take the unperturbed Hamiltonian to be $H_0 = BJ^2$ with $H_0|J, M_J\rangle = E_{JM_J}|J, M_J\rangle$, where the eigen-energy and eigen-function are, respectively, $E_{JM_J} = BJ(J+1)$ and $|J, M_J\rangle$, with spherical harmonics $|J, M_J\rangle$ being functions of $\hat{r}$. The dipole interaction $-DE(\vec{R})\cos\theta_r$ is treated as a perturbation. To first order the wave function becomes

$$\psi_{M_J} = |J, M_J\rangle + \frac{DE}{B}\frac{1}{2(J+1)}\sqrt{\frac{(J+1)^2 - M_J^2}{(2J+1)(2J+3)}}\,|J+1, M_J\rangle - \frac{DE}{B}\frac{1}{2J}\sqrt{\frac{J^2 - M_J^2}{(2J-1)(2J+1)}}\,|J-1, M_J\rangle, \tag{5}$$

and the corresponding eigen-energy to second order in the external E-field becomes

$$\mathcal{V}_{M_J} = BJ(J+1) + \frac{D^2E^2}{B}\frac{1 - 3M_J^2/J(J+1)}{2(2J-1)(2J+3)}. \tag{6}$$

For this model system, M_J is a conserved quantity. Assuming the center of mass motion is adiabatic with respect to the internal state, the above eigen energy from the internal state $\mathcal{V}_{M_J}$ then clearly acts as an effective potential due to its dependence on $\vec{R}$. In particular, we find any state with $1 - 3M_J^2/J(J+1) > 0$ is a weak field seeking state, *i.e.*, free space E-field traps analogous to the B-field IPT can be constructed. These states, of course, are meta-stable, and trapping is possible because of the dynamic stability from the Larmor-like precession of the permanent electric dipole along the direction of a local E-field. On the other hand, the strong field seeking states with $1 - 3M_J^2/J(J+1) < 0$ cannot be used to spatially confine molecules with static fields, because it is impossible to construct a local E-field maximum.

The significant progress gained over the years in trapping and manipulating neutral atoms with static B-fields [12] provide important enabling technologies for the experimental successes of cold atom physics research. A variety of B-field traps have been implemented for various applications [3, 12]. Despite the analogy described earlier of the B-field confinement of magnetic dipoles with the E-field confinement of electric dipoles (of polar molecules), experimental efforts of trapping and manipulating polar molecules with electrostatic E-fields become an active research topic only in recent years with the interests on cold atoms broadening into cold molecules. The Meijer's group pioneered the research of cooling and trapping of molecules with (3D quadrupole) E-fields [13]. They also demonstrated the guiding of cold molecules with a torus shaped (2D) E-field hexapole [14]. With suitable modifications, an analogous IPT of E-field can be constructed for polar molecules [3, 12, 15, 16].

In Fig. 1, we show the calculated dc Stark shift for the $M_J = 1$ state of heteronuclear molecule KRb that is connected to the $J = 2$ manifold when the E-field is absent. We adopt the parameters as from its ground state $X^1\sum \nu = 0$ as measured recently [1] with the electric dipole moment $D = 1.36 \times 10^4\,\mathrm{Hz}\cdot\mathrm{m/V}$ and the rotational constant

$B = 1.1139\,\mathrm{GHz}$. The calculation is accomplished by a numerical diagonalization of the Hamiltonian over a truncated basis of common eigen states for operators $\mathbf{J}^2$ and J_z. The diagonalization is carried out in the subspace of a conserved M_J [10], and the procedure is found to be rapidly convergent. At the E-field strength we consider, the perturbation result ψ_{M_J} in Eq. (5) with $M_J = 1$ and $J = 2$ is found to consists of an excellent approximation. This particular state as illustrated in Fig. 1 is clearly a weak field seeking state. The maximum trap height can become as high as 173 MHz.

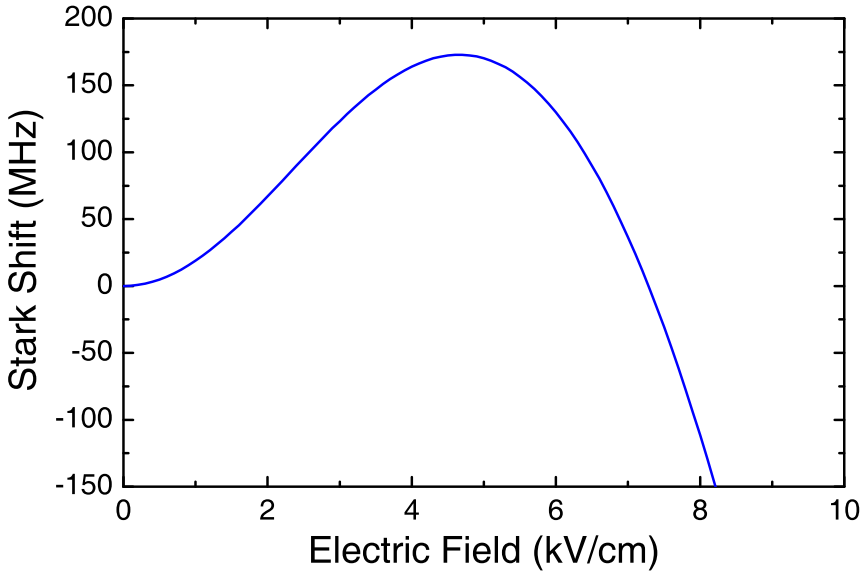

Figure 1. The Stark shift of the $M_J = 1$ state for the polar molecule KRb in the electronic ground state $X^1\sum\nu = 0$, for realistic electrostatic E-trap field strength [16, 15].

3. Rotational transformation of the wave function $|J, M_J\rangle$

Before we extend the Isoshima, *et al.*, protocol [4] from B-field trapped atoms to E-field trapped polar molecules, we will briefly review the rotational properties of the spherical harmonics $|J, M_J\rangle$. Under a rotation along a unit vector $\hat{n}$ by an angle θ, the wave function changes to

$$R(\theta\hat{n})|J, M\rangle = e^{-i\theta\hat{n}\cdot\mathbf{J}}|J, M\rangle. \tag{7}$$

Expressed in terms of the Euler angles, the most general 3D rotation takes the following form

$$R(\alpha, \beta, \gamma) = \exp(-i\alpha J_z)\exp(-i\beta J_y)\exp(-i\gamma J_z). \tag{8}$$

According to the representation theory of rotation, we find

$$\begin{aligned} R(\alpha, \beta, \gamma)|J, M\rangle &= \sum_{M'} D^J_{M'M}(\alpha, \beta, \gamma)|J, M'\rangle \\ &= \sum_{M'} \exp[-iM'\alpha] d^J_{M'M}(\beta) \exp[-iM\gamma]|J, M'\rangle, \end{aligned} \tag{9}$$

where

$$\begin{aligned} d^J_{M'M}(\beta) &= \langle J, M'| \exp[-i\beta J_y]|J, M\rangle \\ &= [(J+M)!(J-M)!(J+M')!(J-M')!]^{1/2} \\ &\quad \times \sum_\nu [(-1)^\nu (J-M'-\nu)!(J+M-\nu)!(\nu+M'-M)!\nu!]^{-1} \\ &\quad \times \left(\cos\frac{\beta}{2}\right)^{2J+M-M'-2\nu} \left(-\sin\frac{\beta}{2}\right)^{M'-M+2\nu}. \end{aligned} \tag{10}$$

The value of ν needs to ensure that the numbers in the factorials stay non-negative. For example, when $\beta = \pi$ and $\cos\frac{\beta}{2} = 0$, for $d^J_{M'M}$ to be nonzero, we need to let $2J + M - M' - 2\nu = 0$. This gives

$$(J - M' + \nu)! = \left[-\frac{1}{2}(M' + M)\right]!, \tag{11}$$

$$(J + M - \nu)! = \left[\frac{1}{2}(M' + M)\right]!. \tag{12}$$

In order to assure that both $J - M' - \nu$ and $J + M - \nu$ are non-negative, we need to make $M' + M = 0$. As a result we find

$$d^J_{M'M}(\pi) = (-1)^{J-M'}\delta_{M',-M}, \tag{13}$$

which gives

$$R(\alpha, \pi, \gamma)|J, M\rangle = (-1)^{J+M} \exp[iM\alpha] \exp[-iM\gamma]|J, -M\rangle. \tag{14}$$

We want to stress at this point that the angles α, β, and γ are parameters used to specify arbitrary rotations, and they have nothing to do with the internal state angles θ_r and ϕ_r. To understand the physics behind the vortex generation protocol, which is directly related to the geometrical properties of the static E-field, we first consider several special cases of unit vectors as rotation axes: for $\hat{n} = \hat{X}\cos\phi + \hat{Y}\sin\phi$, $\hat{n}_\perp = -\hat{X}\sin\phi + \hat{Y}\cos\phi$, and $\hat{n}_q = \hat{X}\sin\phi + \hat{Y}\cos\phi$, respectively, with $\hat{X}$ and $\hat{Y}$ the unit vectors along the Cartesian x- and y-directions. θ and ϕ denote the polar and azimuthal angles of the center of mass coordinate $\vec{R}$. The last case of $\hat{n}_q$ corresponds simply to the direction of a IPT E-field in the plane of $Z = 0$. Using the relationship derived above, we find that

$$\begin{aligned} R_{\hat{n}}(\pi)|J, M_J\rangle &= \exp(-i\pi\hat{n}\cdot\mathbf{J})|J, M_J\rangle \\ &= R_{\hat{z}}(\phi - \pi/2)R_{\hat{y}}(\pi)R_{\hat{z}}(\pi/2 - \phi)|J, M_J\rangle \\ &= -(-1)^{J+M_J}\exp[2iM_J\phi]|J, -M_J\rangle, \end{aligned} \tag{15}$$

$$\begin{aligned} R_{\hat{n}_\perp}(\pi)|J, M_J\rangle &= \exp(-i\pi\hat{n}_\perp\cdot\mathbf{J})|J, M_J\rangle \\ &= R_{\hat{z}}((\pi/2 + \phi) - \pi/2)R_{\hat{y}}(\pi)R_{\hat{z}}(\pi/2 - (\pi/2 + \phi))|J, M_J\rangle \\ &= (-1)^{J+M_J}\exp[2iM_J\phi]|J, -M_J\rangle, \end{aligned} \tag{16}$$

$$\begin{aligned} R_{\hat{n}_q}(\pi)|J, M_J\rangle &= \exp(-i\pi\hat{n}_q\cdot\mathbf{J})|J, M_J\rangle \\ &= R_{\hat{z}}((\pi/2 - \phi) - \pi/2)R_{\hat{y}}(\pi)R_{\hat{z}}(\pi/2 - (\pi/2 - \phi))|J, M_J\rangle \\ &= (-1)^{J+M_J}\exp[-2iM_J\phi]|J, -M_J\rangle. \end{aligned} \tag{17}$$

These properties of rotational transformation show that by enforcing a rotation of the internal state, the wave function gains an appropriate topologically phase specified by the azimuthal angle coordinate of the center of mass (or the molecule) coordinate. Provided the internal state flipping of the permanent dipole moment and the center of mass motion are both adiabatic, the above results show that different vortical phases are generated as in the protocol of Isoshima, *et al.*, for B-field trapped atomic spinor condensates [4, 5, 6].

4. Vortex creation in a condensate of heteronuclear molecules

In this section, we will confirm numerically the vortex creation protocol for a condensate of heteronuclear molecules in an E-field IPT. Basically, we will simulate the axial E-field bias flip and check for the adiabatic conditions of the associated internal state. There are two main points we need to watch for. First we need to create the proper vortical phase structure on a condensate during the time evolution of the E-field. Secondly we must maintain adiabaticity during the flip of the internal state of a polar molecule.

Considering the first question, assume the initial state corresponds to a polar molecule locally aligned along the E-field direction of a E-field IPT, which is essentially along the axial z-axis direction close to the z-axis, its local internal state $\phi_{M_J}(\theta_r, \phi_r)$ can then be described approximately as the eigen state of the system in the local E-field. Since M_J is a conserved quantity, this eigen state $\psi_{M_J}(\theta_r, \phi_r)$ can be expanded by a series of $|J, M_J\rangle$ in J,

$$\psi_{M_J}(\theta_r, \phi_r) = \sum_J C_J |J, M_J\rangle. \tag{18}$$

The flipping of the E-field bias adiabatically corresponds then to nothing but a rotation of the initial state along the unit vector $\hat{n}_q(\theta, \phi)$ in the transverse plane by an angle of π. According to the rotation properties we discussed before, this flipping of the bias then gives

$$R_{\hat{n}_q}(\pi)\psi_{M_J}(\theta_r, \phi_r) = \sum_J (-1)^{J+M_J} C_J \exp[-2iM_J\phi]|J, -M_J\rangle, \tag{19}$$

which gains clearly a vortex with a winding number $-2M_J$.

Turning to the second question concerning the adiabaticity, we simply can propagate the initial wave function ψ_{M_J} in real time, simulating the complete process of the bias flip. To test the level of adiabaticity and the validity of Eq. (19), we consider points $(X_0, Y_0, 0)$ along a circle in the X-Y plane. The absolute value for the E-field along the circle is then a constant, although their local directions are all different. In fact, for the E-field IPT being considered here, the E-field can be written as $\vec{E} = E'(X, -Y, L)$ with L a constant. Choosing the weak field seeking state $\psi_{M_J=1}$ with $J = 2$ at $\vec{E} = 3\,\mathrm{kV/cm}\,\hat{\mathrm{Z}}$ as considered in Fig. 1, we simulate the flip protocol over various time intervals. The time evolution of the bias E-field is taken to be the same as that in the optimal B-field protocol [9], and the E-field in the transverse X-Y plane is treated as a constant.

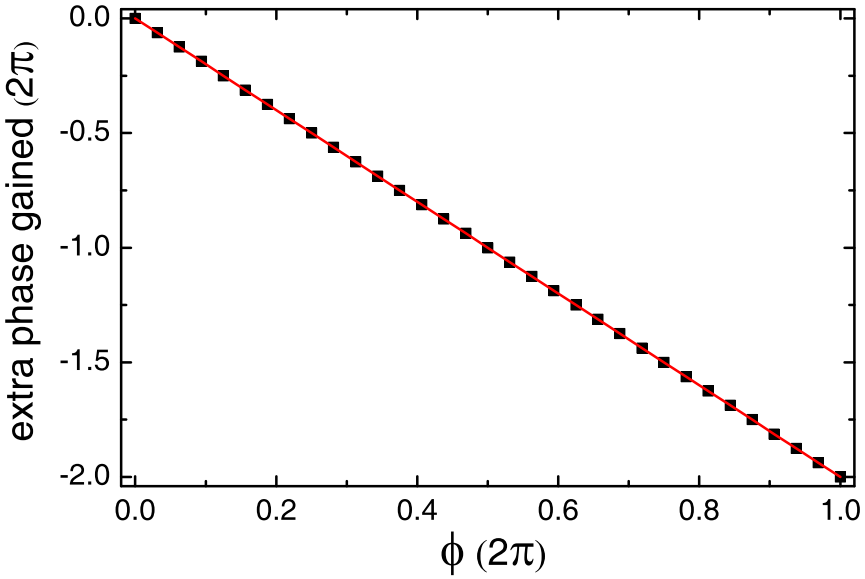

Figure 2. The topological phase gained after numerically flipping the bias E-field, that is calculated by taking the inner product of the time evolved state after flip with the nominally down polarized ($M_J = -1$) state. ■ denotes numerically computed data, and the red solid curve represents a linear fit. Both axes are dimensionless in units of 2π.

In our simulation, we take the E-field at $t = 0$ as $E'L = 3\,\mathrm{kV/cm}$ and $E'\sqrt{X_0^2 + Y_0^2} = 0.3\,\mathrm{kV/cm}$. After the flip of the E-bias over $10^5/B$ ($\sim$ 0.1 ms at $B = 1.1139\,\mathrm{GHz}$), which is a reasonably fast time scale, we calculate the inner product of the final state with the eigen-state $\psi_{M_J=-1}$ (of $J = 2$) when the E-field $\vec{E} = -3\,\mathrm{kV/cm}\,\hat{\mathrm{Z}}$ is pointed in the downward direction. Using these parameters, the adiabaticity is found to be fully satisfied, and the state overlap is essentially 100% except for the extra topological phase gained as shown in Fig. 2. A numerical fit gives precisely the slope for the phase over ϕ being exactly equal to -2, which confirms the high fidelity operation of our vortex pump proposal.

Before concluding, we point out that, like the system of magnetically trapped atomic spinors inside a B-field IPT [17, 9], the quantity of $J_z - L_z$ is found to commute with the Hamiltonian $B\mathbf{J}^2 - \mathbf{D} \cdot E'(X, -Y, L)$, where E' is the spatial gradient of the E-field IPT, and L_z is the mechanical angular momentum of the heteronuclear molecule. $\vec{J}$ is the rotational angular momentum of the molecule. If the flip of the E-bias is indeed adiabatic, the respective quantum numbers conserve the combination $M_J - L_z$. After going through the flip, the internal rotational state is changed from M_J to $-M_J$, which then must be accompanied by an increase of $2M_J$ to its mechanical angular momentum. Thus we see the appearance of a vortex state.

In conclusion, by analogy with B-field trapping of neutral atoms with magnetic dipoles, we study and identify weak field trapping states of polar molecules inside a spatially inhomogeneous dc E-field. Further, we suggest that an effective and efficient vortex pump protocol can be envisioned for condensates of polar molecules [8, 9],

based on the flipping of the axial bias field, originally suggested [4] and experimentally demonstrated for atomic spinor condensates inside a B-field IPT [5, 6, 7]. We have confirmed that the E-field bias flip remains effective, and the vortex state created maintains a vorticity proportional to the M_J quantum number of the trapped molecule state. When a diatomic molecule is placed inside a homogeneous E-field, M_J is a good quantum number. The weak field trapping states can be associated with very large values of M_J, thus the amount of vorticity created could become very significant even after a single bias flip. This vorticity possibly could open up a practical approach to reach the rapid rotation limit of atomic/molecular quantum gases.

More generally, we find that this protocol for vortex creation remains effective if the diatomic molecule is taken as a symmetric top [18]. Interestingly we find the E-bias flip also works for the strong field trapping states provided the polar molecules are confined through other means not relying on its permanent electric dipole. Finally, other improvements to the B-field bias flip protocol [4], such as those developed for cyclically operated continuous vortex pumping schemes can be analogously extended to the case of heteronuclear molecules [8, 9].

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